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# BCC vs. HCP - The Effect of Crystal Symmetry on the High Temperature Mobility of Solid $^4\text{He}$

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**Abstract** We report results of torsional oscillator (TO) experiments on solid  $^4\text{He}$  at temperatures above 1K. We have previously found that single crystals, once disordered, show some mobility (decoupled mass) even at these rather high temperatures. The decoupled mass fraction with single crystals is typically 20- 30%. In the present work we performed similar measurements on polycrystalline solid samples. The decoupled mass with polycrystals is much smaller,  $\sim 1\%$ , similar to what is observed by other groups. In particular, we compared the properties of samples grown with the TO's rotation axis at different orientations with respect to gravity. We found that the decoupled mass fraction of bcc samples is independent of the angle between the rotation axis and gravity. In contrast, hcp samples showed a significant difference in the fraction of decoupled mass as the angle between the rotation axis and gravity was varied between zero and 85 degrees. Dislocation dynamics in the solid offers one possible explanation of this anisotropy.

## 1 Introduction

The physical mechanism responsible for the apparent mobility of solid He remains a subject of intense study. Experiments done on solid  $^4\text{He}$  contained inside a torsional oscillator (TO) show a partial mass decoupling<sup>1,2,3,4,5,6,7</sup> below some 200 mK, an indication of some kind of mobility of the solid. The mass decoupling fraction seen in the various TO experiments is typically 0.01%-2%, except

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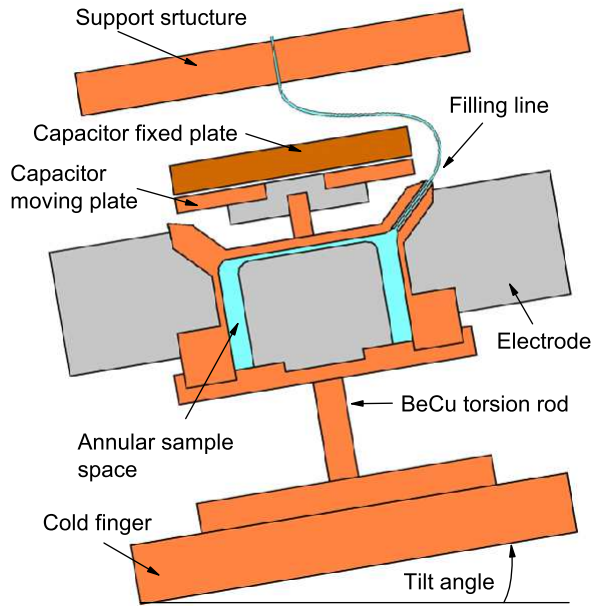
for the unique results of 20% by Rittner and Reppy<sup>2</sup>. The interpretation of these results is under debate in terms of several competing models. These include supersolidity<sup>8,9</sup>, superglass<sup>10,11,12</sup>, and dislocation mediated effects<sup>13,14,15,16</sup>. The possibility of dislocation dynamics came up following measurements of the shear modulus which showed changes at the same temperature where mass decoupling was observed<sup>17,18</sup>.

Our previous contribution to this subject came through TO measurements on solid He at higher temperatures, between 1.1K and 1.9K. In contrast to polycrystalline samples used by others, we grew single crystals inside the sample space of the TO. Crystals of commercially pure <sup>4</sup>He or of 100 ppm <sup>3</sup>He-<sup>4</sup>He mixtures were grown at a constant temperature and pressure. We found that generation of structural disorder within a single crystal caused a large fraction of the mass to decouple from the TO<sup>19,20</sup>. The decoupled mass fraction did not depend strongly on temperature. The very fact that mass decoupling in a TO experiment can be observed at practically any temperature above 1K suggests that the phenomenon is not an usual phase transition.

One way to distinguish between the physical scenarios mentioned above is to look for some signs of anisotropy. The mass decoupling effect is seen in the response of the solid to stress applied by the moving wall of the cell. Supersolidity or glassy models of the solid do not predict any anisotropy of this response. In contrast, if the response is due to dislocations, one should expect some anisotropy. For example, at small values of externally applied stress, it is reasonable to assume that dislocations glide. Glide occurs on selected crystalline planes, and is naturally sensitive to the direction of stress relative to the crystalline axes. In order to check whether this is the case, one needs to vary the orientation of the crystal with respect to that of the stress. Since the stress is applied by the walls of the TO, one should grow crystals at different orientations relative to the rotation axis of the TO. The experiment described here was designed to test this scenario.

## 2 Experimental System

The interaction of He atoms with practically any substrate is much stronger than the He-He interaction. This has important consequences on the growth of He crystals. The strong interaction with the substrate causes the typical substrate (inner walls of the cell in our case) to be coated by a dense, close packed layer of solid He. Consequently, He crystals always grow on a substrate consisting of amorphous He and are not sensitive to the material from which the cell is constructed. The orientation of the crystal is determined by gravity and by the thermal profile inside the cell. As a result, Helium crystals tend to grow more or less with the same crystalline orientation in relation to gravity. For example, in X-ray studies done by Greywall<sup>21</sup>, it was found that 80% of 99 hcp crystals grown had their c axis at an angle of less than 30° from the horizontal direction. Similar behavior was observed in neutron scattering experiments<sup>22,23</sup>, with the c axis of hcp crystals again being close to horizontal. Regarding bcc crystals, we found that these typically grow with the [111] direction close to vertical<sup>22,24,23</sup>. Therefore, experiments on different crystals in the same cell are usually quite reproducible, as most of these crystals would grow with the same crystallographic direction with respect to the cell. In order to measure the decoupled mass fraction of crystals grown with



**Fig. 1** Schematic cross section of the torsional oscillator (TO) assembly. The drawing is to-scale. The assembly can be tilted between zero and 90 degrees, in order to grow crystals at different angles with regards to gravity. Note that the filling line enters the cell at a corner so that the entrance is always at the highest point of the cell. This prevents the filling line from becoming blocked with solid before the cell is full, irrespective of the tilt angle of the TO. The bottom of the cell is thermally connected to a  $^3\text{He}$  refrigerator. The crystals are grown in the annular space of outer diameter of 18mm.

different crystalline directions, one has to change the direction of the stress relative to the crystalline axes. Since the orientation of the crystals tends to remain fixed in space, one has to change the orientation of the torsional oscillator (TO). To that end, we built a TO cell which could be tilted, so that the rotation axis was not parallel to gravity. We grew the crystals by raising the pressure in the cell, using an open and slightly heated filling line. One additional requirement was that the filling line must enter the cell at the highest point, so that it will not become blocked with solid before the cell is full. For that, we designed the cell with the filling line entering it from the top corner. Figure 1 shows an in-scale schematic cross section of the cell. We grew bcc and hcp solids in this cell and tested the mass decoupling with the cell's rotation axis aligned with the direction of gravity, and with the rotation axis forming an angle of 2, 5, and 85 degrees with the direction of gravity.

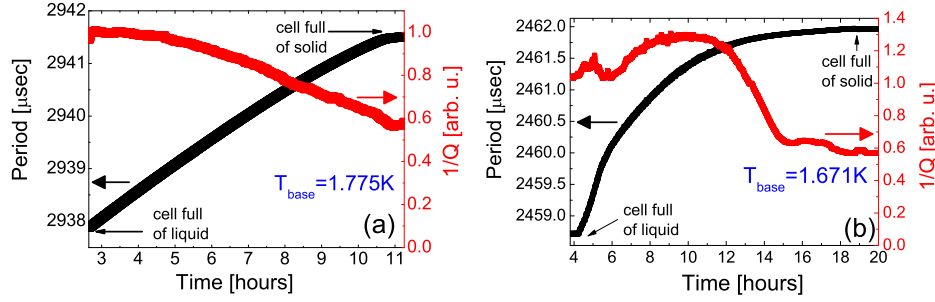
The cell itself was made of beryllium copper with a Stycast bob in its center, which defines an annular sample space of 11 mm height and 2 mm width. The internal volume was  $1.2 \text{ cm}^3$ . Solid He was grown inside this annular space. All the internal corners were rounded, with sharp corners remaining only at the bottom of the cell. We used a capacitive pressure gauge to measure the pressure in situ. The gauge, seen in figure 1, includes one (moving) capacitor plate attached to the top of the cell and another (fixed) plate connected to an external support structure.

In this study, we grew a total of 21 crystals at temperatures between 1.39K and 1.75K. Most of the samples were grown using commercially pure  $^4\text{He}$ , but we also grew some samples containing 100ppm of  $^3\text{He}$ . Initially, the cell's resonant period was  $2461 \mu\text{sec}$ . In order to check whether any of the results depend on the frequency at which the oscillator operates, we changed the resonant period of the cell from  $2461 \mu\text{sec}$  to  $4388 \mu\text{sec}$  by reducing the diameter of the torsion rod. In order to eliminate any possible influence of the oscillatory motion of the TO on crystal growth, several crystals were grown with the TO turned off. All these samples showed the same mass decoupling fraction as the samples grown with the TO oscillating.

### 3 Results

The solid He samples grown in the TO cell were all polycrystalline. The likely reason for that is the temperature profile inside the cell, which was not symmetric about the rotation axis of the TO. The hottest point in the cell was the entry point of the heated filling line, which was off-center for reasons explained above. For comparison, in panel (a) of figure 2 we show the time dependence of the period and dissipation of the TO during growth of a single crystal. These data were taken with the cell used in our previous work<sup>20</sup> in which the rotation axis was parallel to gravity, and the filling line entered the cell at the top. The approximately linear increase of the period with time is consistent with the crystal growing from the bottom of the cell, and gradually filling the sample space with the liquid-solid interface horizontal. During the growth, the dissipation of the TO decreases continuously, indicating that the crystal is of good quality with low internal friction. Turning to panel (b) of figure 2, here we show similar data for a TO rotated at 85 degrees. These data were taken during the current experiment. The dependence of the period on time is consistent with the solid growing around the circumference of the sample space, filling the cell from the outside towards the center. At the beginning of the growth, the dissipation of the TO increases. This indicates that many solid grains are created, and there is internal friction between them. Obviously, this type of growth results in a polycrystal. The resonant period change upon growth in the tilted cell was  $3.25 \mu\text{sec}$ , about the change expected from the classical moment of inertia. For completeness, we mention that the solid in both experiments shown in figure 2 was grown by applying a small constant overpressure.

At the end of the growth, the entrance to the cell becomes blocked with solid and the temperature profile changes abruptly. This causes stress, and leads to additional structural disorder in the sample. As before<sup>19</sup>, we observed that some fraction of the mass decoupled from the TO. All the polycrystalline samples showed some mass decoupling, the fraction ranging between 0.1% and 1.3%. The initial



**Fig. 2** Time dependence of the period and dissipation of the TO during solid growth. The black symbols in both plots are the resonant period of the TO, which at the beginning is that of a cell with no solid, and at the end of the growth process is that of a cell full of solid helium. The red symbols show the dissipation during this process. Panel (a) shows the period and dissipation during growth of a single crystal in a TO aligned with gravity which has a filling line entering it from the top. In this case, the solid grows as a single crystal. Panel (b) shows the period and dissipation of a TO tilted at an angle of 85 degrees with respect to the direction of gravity. The time dependence of the period in (b) is consistent with a polycrystalline growth.

mass decoupling fraction depended on the intensity of the pressure/temperature step which disordered the solid. After cooling the TO, the mass decoupling fraction reached a limiting value for each direction and crystal symmetry.

For the bcc phase, this limiting value was around 1.3%, independent of the orientation of the TO. For the hcp phase, there were differences between samples grown with different orientations of the TO. Hcp samples grown with the rotation axis aligned with gravity, or tilted at an angle of at most 5 degrees, reached the same mass decoupling fraction as the bcc crystals. Hcp crystals grown in the TO tilted at an angle 85 degrees reached a limiting value of 0.75% - about half of the value of the bcc phase. These values are the typical results obtained with 21 different crystals, 15 of them grown as bcc (2 grown at 0 degrees tilt, 3 at 2 degrees, 5 at 5 degrees, and 5 at 85 degrees), and 6 grown as hcp (3 at 5 degrees and 3 at 85 degrees tilt). There was no apparent difference between the 0, 2 and 5 degree tilts, and only the 85 degree tilt gave a significant difference between the hcp and bcc crystals.

We found that the size of the mass decoupling was the same for crystals grown using commercially pure  $^4\text{He}$  and for those grown using a mixture containing 100ppm of  $^3\text{He}$ . Similarly, we found that the results did not depend on the period of the TO.

Comparing these mass decoupling values to those obtained in our previous experiments<sup>19,20</sup> using single crystals, we see a huge difference. Except for the solid being polycrystalline in the current experiment and a single crystal in our previous work, we used the same cryostat, materials and growth methods in both experiments. Therefore, it seems that polycrystallinity is the cause of the difference in

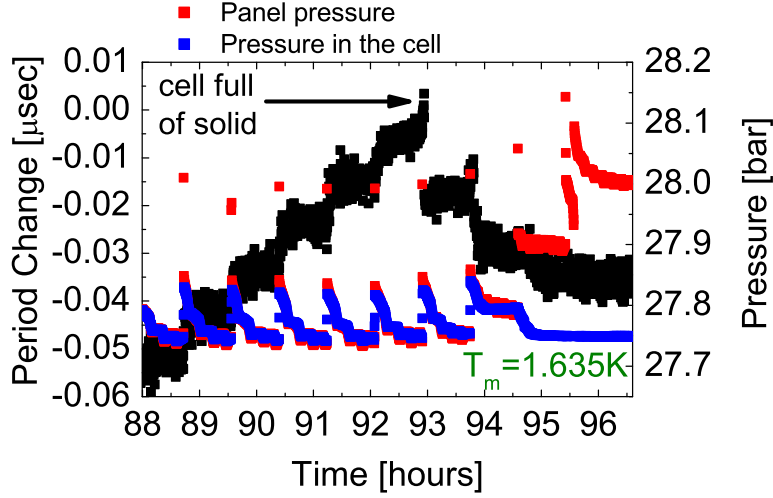
decoupled mass. We mention that the decoupled mass fraction with polycrystals is very similar to the one seen in low temperature experiments using  $^4\text{He}$  of commercial purity<sup>1,2,3,4,5,6,7</sup>.

To reduce the influence of the initial disordering process, we grew bcc samples and then cooled them through the bcc-hcp transition into the hcp phase. Figures 3 and 5 show such data. In this case the solid was grown by periodically adding small amounts of He to the cell. Figure 3 shows the final stages of the growth of a bcc solid. Once the cell is full (at 93 hours), mass decoupling takes place and the period of the TO decreases. This mass decoupling was produced by applying several pressure pulses to a cell filled with solid. The first pressure pulse after the cell is completely full of solid only begins solidifying the helium in the filling line, so the pressure is still transmitted into the cell. The following pressure pulses are with the filling line progressively blocked with solid. These pulses cannot change the pressure inside the cell directly, but can still cause stress due to change of the heat flux into the cell, changing the temperature profile within. At this stage, the sample gradually cools to the set temperature of the TO stage. This is the reason why the period of the TO continues to decrease even after the filling line is blocked. This is evident from the pressure in the cell. As can be seen in the figure, after 94.5 hours the filling line is blocked, and subsequent external pressure pulses no longer influence the pressure inside the cell.

Panel (b) of figure 4 shows the what happens once this sample is cooled. The bcc solid gradually converts into the hcp phase during cooldown. It is seen that the decoupled mass fraction decreases during this stage, and vanishes at the lower triple point of 1.46K. We remark that bcc crystals grown at temperatures less than 1.7K, inevitably pass through the lower triple point during cooldown. At the triple point, the bcc to hcp conversion is done in the presence of liquid. This transformation occurs spontaneously inside an isolated cell (the filling line is blocked with solid). If there is any mass decoupling associated with this process, it is independent of external factors.

In figure 5 we show the spontaneous disordering of the hcp solid created this way, after the cell was cooled further to 1.285K. As the sample cooled, the fluid in the cell gradually solidified. At 1.285K, the cell was full of hcp solid which was then disordered due to thermal stress and showed mass decoupling. This sequence of events took place irrespective of the orientation of the TO. We can therefore compare the size of the mass decoupling in the bcc and hcp phases, for crystals that underwent the same procedure, but were grown with the TO at different orientations. When the TO's rotation axis was aligned with the direction of gravity, or at most 5 degrees from it, the bcc phase and the hcp phase showed the same maximal mass decoupling value within experimental error. On the other hand, when the rotation axis was at an angle of 85 degrees from the direction of gravity, the bcc solid reached a maximal mass decoupling value of around 1%, whereas the hcp reached only 0.1%, as shown in figure 5.

Figure 4 shows the temperature dependence of the mass decoupling of two different crystals grown in a cell tilted at an angle of 85 degrees. As can be seen, the hcp crystal reaches much lower values than the bcc one, even after significant cooling. The results for the 0-5 degree tilt give the same temperature dependence reported in our previous work<sup>20</sup>, except for the values of the decoupled mass being much smaller.



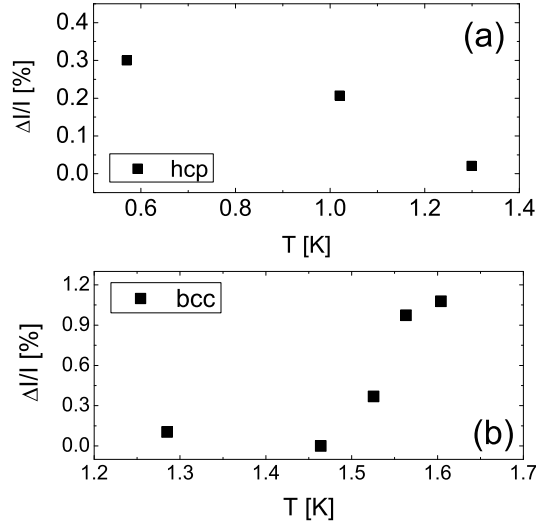
**Fig. 3** Mass decoupling of a solid sample grown as bcc. In this experiment the TO's rotation axis was tilted by 85 degrees with respect to gravity. The figure shows the final stages of the growth process, followed by mass decoupling (decrease of the period). The decoupled mass fraction reached a constant value of 1% after several hours of relaxation. Red and blue symbols show the externally applied pressure and the pressure inside the cell respectively.

#### 4 Discussion

The strong dependence of the decoupled mass fraction on the growth direction in the anisotropic hcp crystal might be an indication that dislocations play an important role in the phenomenon. Bcc crystals are isotropic and there are many ( $\{110\}$ ,  $\{112\}$ , and  $\{123\}$ ) equivalent slip planes. If the mass decoupling effect is associated with dislocation glide, we expect that in the bcc solid the coupling to the TO will show little dependence on the angle between the rotation axis and the crystallographic direction. In the hcp crystal, on the other hand, there is only one easy slip plane for edge dislocations, (0001). Therefore, dislocations would glide easily in the bcc structure no matter what the direction of stress is, while in the hcp structure the glide will be much more sensitive to the direction of stress in space, which is determined by the orientation of the cell.

The small amount of decoupled mass seen in our experiment using polycrystalline samples is similar to that observed in the low temperature TO experiments on solid helium<sup>25,18,2,3,4,5,6</sup>. In the low temperature experiments the crystals were grown using a blocked capillary method which results in polycrystalline samples<sup>26</sup>, with the exception of one experiment<sup>27</sup>, in which the cell contained sharp corners, which again makes it impossible to fill it with a single crystal.

To conclude, we found that polycrystals grown as bcc always gave the same mass decoupling fraction, regardless of the growth direction. Hcp polycrystals, on

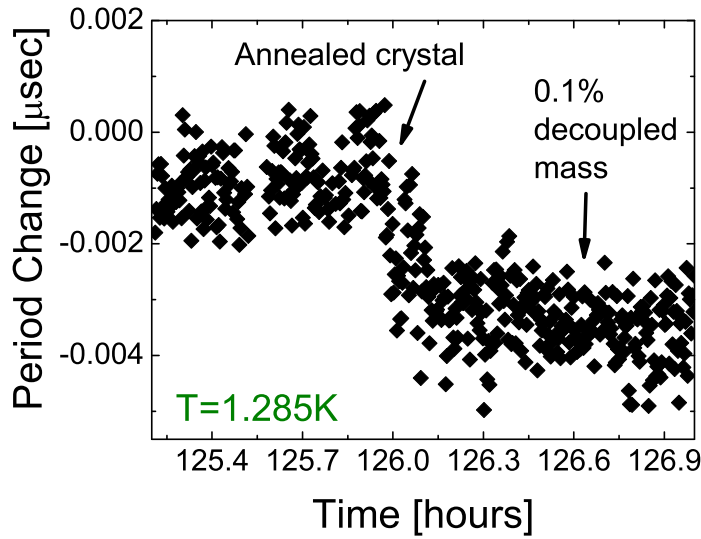


**Fig. 4** Temperature dependence of the mass decoupling for two samples grown in a TO with a rotation axis tilted by 85 degrees with respect to gravity. Panel (a) an hcp solid grown at 1.419K, and panel (b) a bcc solid grown at 1.635K, cooled through the bcc-hcp coexistence region down into the hcp phase.

the other hand, showed much smaller mass decoupling values when grown perpendicular to the direction of the rotation axis. Dislocation glide could be responsible for the apparent anisotropy associated with the mass decoupling inside the TO.

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**Fig. 5** Spontaneous mass decoupling of an hcp solid sample. This sample was initially grown as bcc and then cooled through the triple point to the hcp phase. The TO's rotation axis was tilted by 85 degrees. Upon further cooling of the solid shown in figure 3, as it reached the lower triple point, it annealed and the decoupled mass fraction became zero. The crystal followed the melting line down to a temperature of around 1.285K, where the cell became full of solid, and spontaneous mass decoupling took place. In this case, the decoupled mass fraction reached a much lower value, of 0.1%. Note that the vertical scale in this figure is about a factor of 10 finer than that in figure 3. This illustrates the different behavior of bcc and hcp solids in the tilted TO.

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